Implementing universal multi-qubit quantum logic gates in three and four-spin systems at room temperature

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In this paper, we present the experimental realization of multi-qubit gates Λ_n (*not*) in macroscopic ensemble of three-qubit and four-qubit molecules. Instead of depending heavily on the two-bit universal gate, which served as the basic quantum operation in quantum computing, we use pulses of well-defined frequency and length that simultaneously apply to all qubits in a quantum register. It appears that this method is experimentally convenient when this procedure is extended to more qubits on some quantum computation, and it can also be used in other physical systems.

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I. INTRODUCTION

A quantum computer, exploiting quantum state superposition and entanglement, is capable of performing select computations more powerfully than any classical computer [1,2]. Many quantum algorithms that achieve the same computational tasks much faster than their classical counterparts have been designed [3–5]. These theoretical results have led many groups to try to realize a quantum computer experimentally. Up to now, there are a rapidly growing number of proposed device technologies for quantum computation [6–9]. Of these technologies, those exploiting nuclear magnetic resonance(NMR) have been the first to demonstrate non-trivial quantum algorithms with small numbers of qubits [11–13], most authors realized three-qubit NMR quantum computation [14–18].

One of the key challenges we are facing is to increase the size of the system used. It is notable that the largest number of qubits used up to date is seven-qubit [10]. To experimenter, any computational task could be viewed as a quantum circuit composed of quantum logic gates [19], in analogy to the situation of classical computers. In addition, from the point of view of decoherence, the circuit must be high efficient in experiment. Therefore, it is important to demonstrate the Multi-qubit quantum logic gates on a large number of qubits effectively in practice. On the other hand, some methods to construct the arbitrary multi-qubit quantum circuit with a sequence of universal two-qubit gates were reported recently [20–22].

In this paper, we start from the introduction of Λ_n (*not*) gate, and try to realize it with a sequence of two-qubit Λ_1 (*not*) gates presented by Barenco et al [20]. Through this procedure we find that in NMR doing nothing (suspending evolution) in one part of a system while doing something in another part of the system is nontrivial, i.e. the extension of two-qubit gate to more than two coupled spins is complicated and not easy to implement. Then, we report the implementation of Λ_2 (*not*) and Λ_3 (*not*) gates by utilizing the electromagnetic pulses of well-defined frequency and length in three and four-qubit systems. For the sake of experimental convenience, these pulses are physically reasonable choice for the method of quantum circuit construction in quantum computation rather than two-qubit Λ_1 (*not*) gate. It seems that this method is experimentally convenient when we extend this procedure to more qubits, and one can perform a large class of multi-qubit controlled rotations simultaneously in many physical systems such as quantum dots, nuclear spins and trapped ions *etc.*

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II. QUANTUM $\Lambda_N (NOT)$ GATE

The $\Lambda_n (not)$ gate is a $(n+1)$-qubit quantum logic gate in which there are n control qubits and one target qubit, performing the unitary transformation

$$\Lambda_n (not) (|x_1, ..., x_n, y\rangle) = \left| x_1, ..., x_n, \bigwedge_{k=1}^{n} x_k \oplus y \right\rangle \tag{1}$$

$$= \begin{cases} |x_1, ..., x_n, y\rangle & \text{if} \bigwedge_{k=1}^{n} x_k = 0 \\ |x_1, ..., x_n, \overline{y}\rangle & \text{if} \bigwedge_{k=1}^{n} x_k = 1 \end{cases}$$

for all $x_1, ..., x_n, y \in \{0, 1\}$, and $\bigwedge_{k=1}^{n} x_k$ denotes the AND of the boolean variables $\{x_k\}$. It can be seen that n control qubits does not change their values $|x_1, ..., x_n\rangle$ after the action of the $\Lambda_n (not)$ gate and the target qubit flips its state $|y\rangle$ to $|\overline{y}\rangle$ if and only if $\bigwedge_{k=1}^{n} x_k = 1$. The $\Lambda_n (not)$ gate can also be described by the following unitary matrix

$$\Lambda_n (not) = \begin{pmatrix} I_{2^{n+1}-2} & 0 \\ 0 & \sigma_x \end{pmatrix} \tag{2}$$

where the basis states of $n+1$ qubits are lexicographically ordered, i.e., $|0 \cdots 00\rangle, |0 \cdots 01\rangle, \cdots |1 \cdots 11\rangle$, $I_{2^{n+1}-2}$ is the $(2^{n+1} - 2) \times (2^{n+1} - 2)$ identity matrix and σ_x are Pauli spin matrices.

when $n = 1$, $\Lambda_1 (not)$ is the so-called CNOT gate

$$\Lambda_1 (not) = |00\rangle \langle 00| + |01\rangle \langle 01| + |10\rangle \langle 11| + |11\rangle \langle 10| = \begin{pmatrix} I_2 & 0 \\ 0 & \sigma_x \end{pmatrix} \tag{3}$$

It has a role to play in quantum measurement, in creation and manipulation of entanglement and in quantum error correction. Furthermore, $\Lambda_1 (not)$ gate is of central importance in quantum computation because any quantum logic gates can be decomposed into a sequence of one-qubit rotations and two-qubit $\Lambda_1 (not)$ gates [20]. Most implementation of the $\Lambda_1 (not)$ gate in a liquid state NMR involved a special sequence of resonance electromagnetic pulses. For example, in two-spin physical system of the carbon-13 labeled chloroform, the "modified" $\Lambda_1 (not)$ gate was implemented using two radio-frequency pulses [23]. The first pulse induced a $\pi/2$ rotation of the target spin around the one axis of the rotating reference frame. The second pulse induced a similar rotation around the other axis. The delay time between two pulses was $\pi/2J$, where J is the interaction constant. This action is modified in the sense that it differs from $\Lambda_1 (not)$ gate in relative phases but it require even less resources. One can also see the ideal implementation of $\Lambda_1 (not)$ gate with more resources [24,25].

If $n = 2$, $\Lambda_2 (not)$ can be regarded as a three-qubit quantum logic gate which is named as Toffolli-gate now, and this gate plays the role of a universal gate for reversible circuits.

$$\Lambda_2 (not) = \sum_{x=000}^{101} |x\rangle \langle x| + |110\rangle \langle 111| + |111\rangle \langle 110| = \begin{pmatrix} I_6 & 0 \\ 0 & \sigma_x \end{pmatrix} \tag{4}$$

Similarly, the four-qubit gate $\Lambda_3 (not)$ can be written as

$$\Lambda_3 (not) = \sum_{x=0000}^{1101} |x\rangle \langle x| + |1110\rangle \langle 1111| + |1111\rangle \langle 1110| = \begin{pmatrix} I_{14} & 0 \\ 0 & \sigma_x \end{pmatrix} \tag{5}$$

One method to implement $\Lambda_2 (not)$ gate and $\Lambda_3 (not)$ gate would be to make use of one-qubit rotations and two-qubit $\Lambda_1 (not)$ gates. several schemes have been proposed to realize $\Lambda_n (not)$ gates by these foundational quantum logic gates. However, extending the number of qubits has not been proved easy. Barenco et al. exhibited to obtain the best known three-qubit Toffoli gate [26]($\Lambda_2 (not)$) with five $\Lambda_1 (not)$ gates; in this same paper, they also built up a $\Lambda_3 (not)$ gate with thirteen $\Lambda_1 (not)$ gates [20]. Reck *et al.* proved that any n-qubit quantum circuit, expressed as an unitary operator, could be constructed using a finite number $(\Theta(n^3 4^n))$ of two-qubit gates [27]. More important, due to the interactions among the qubits, to apply two-qubit gate while doing nothing on the other $(n-2)$ qubits in n-qubit system is much more complicated than to apply the same two-qubit gate in a two-qubit NMR quantum computer [28].

The above scheme to realize the $\Lambda_n (not)$ gates $(n \geq 2)$ depends heavily on using the fundamental two-qubit $\Lambda_1 (not)$ gate. Although each gate in these schemes can be implemented by appropriately refocused evolutions in NMR,

however, the approach to realize $\Lambda_n \, (not)$ gate in the n-qubit NMR system according to these theoretical schemes is not efficient in general. There are two main reasons. One is that most of these schemes are to construct the arbitrary quantum circuit with a sequence of the two-qubit operation $\Lambda_1 \, (not)$. This may not be a physically reasonable choice in some NMR quantum computation, but for the moment this should be considered as a mathematical convenience which will permit us to address somewhat general questions. The other is that NMR quantum operations depend heavily on the molecule being used. The fact that the chemical shift and coupling constant are small in the spin systems leads to the requirement of the highly frequency selectivity which is important in quantum operation.

III. EXPERIMENTAL REALIZATION

Seth Lloyd described a method to perform arbitrary quantum circuit by a sequence of electromagnetic pulses of well-defined frequency and length in a weakly-coupled quantum systems [29,30]. For the purpose of interpreting this method, let us consider a macroscopic ensemble of n-spin molecules at room temperature. The n nuclear spins in each molecule represent a n-qubit register. The qubits are labeled by the characteristic frequencies, ω_k, ($k = 1$ to n) due to the Zeeman interaction of the nuclear spins with the magnetic field. The Hamiltonian of the nuclear spins in a molecule in solution is well approximated for the following reason: small interactions with other nuclei do not play a major role in the dynamics while higher order terms in the spin-spin coupling can be disregarded in the first-order model, and the rapid molecular tumbling averages away in the liquid at a high magnetic field.

$$H_{n \; qubits} = -\frac{\hbar}{2} \left[\sum_{k=1}^{n} \omega_k \sigma_z^k + \pi \sum_{k>m} J_{m,k} \sigma_z^m \sigma_z^k \right] \tag{6}$$

where ω_k is the Lamour frequency of spin k and σ_z^k is the $\widehat{z}$ Pauli operator of spin k. $J_{m,k}$ is the strength of scalar weakly coupling between k spin and m spin. Because the energy level differences are small at room temperature $E_k/K_B T \ll 1$ and scalar coupling is weak $J \ll |\omega_{k+1} - \omega_k|$; the initial state is the thermal equilibrium state,

$$\rho_{eq} = \frac{\varrho^{-E_k/K_B T}}{\sum_{k=0}^{2^n+1} \varrho^{-E_k/K_B T}} \tag{7}$$

we use the complete set of the basis states, $\{ |0_1 \cdots 0_{n-1} 0_n \rangle = |0\rangle , |0_1 \cdots 0_{n-1} 1_n \rangle = |1\rangle , \cdots, |1_1 \cdots 1_{n-1} 1_n \rangle = |2^n - 1\rangle \}$, form a complete set of eigenstates of Hamiltonian H: $H |k\rangle = E_k |k\rangle$, ($k = 0, \cdots, 2^n - 1$). For example, the energies of the ground state, $|0 \cdots 00\rangle$, and the excited state, $|1 \cdots 11\rangle$, are

$$E_0 = -\frac{\hbar}{2} \sum_{k=1}^{n} \left(\omega_k + \pi \sum_{k>m} J_{m,k} \right) \tag{8}$$

$$E_{2^n-1} = -\frac{\hbar}{2} \sum_{\alpha=1}^{n} \left(-\omega_k + \pi \sum_{k>m} J_{m,k} \right)$$

we assume that: $\omega_1 < \omega_2 < \omega_3, \cdots, < \omega_n$, and $J \ll \omega_{k+1} - \omega_k$, ($k = 1, 2, \cdots, 2^n - 1$).

Assume that the frequency of the external magnetic field, ω, is resonant with the frequency of the transition, $|1_1 \cdots 1_{n-1} 0_n \rangle \leftrightarrow |1_1 \cdots 1_{n-1} 1_n \rangle$, and that this transition frequency is different from all other single-spin transition frequencies. In this case, by applying a single π-pulse with frequency ω, we could implement the modified quantum $\Lambda_{n-1} \, (not)$ gate in an ensemble of n-spin molecules,

$$U = \varrho^{-i\pi \left[\frac{1}{16} \left(1-\sigma_z^1\right), \cdots, \left(1-\sigma_z^{n-2}\right) \left(1-\sigma_z^{n-1}\right) \sigma_x^n \right]} = \begin{pmatrix} I_{2^n-2} & 0 \\ 0 & -i\sigma_x \end{pmatrix} \tag{9}$$

The above operation changes the state of the n-th spin (the target spin): $|0\rangle \leftrightarrow -i |1\rangle$, only if the state of any other spins is $|1\rangle$. Some groups tested this method experimentally in their small two-qubit and three-qubit NMR quantum computers [17,18,31]. Here, we extend this method to four-qubit systems and realized the Toffoli gate and $\Lambda_3 \, (not)$ gate experimentally.

A. three-qubit Toffoli gate

The single-pulse experimental realization of a modified three-qubit quantum $\Lambda_2\,(not)$ gate (Toffoli gate) is shown in Figure 1. In our NMR approach to realize three-qubit $\Lambda_2\,(not)$ gate we need three qubits. A convenient system we used is the liquid consisting of identical chlorostyrene molecules, the three weakly coupled spin 1/2 hydrogen nuclei, which are distinguished by their different resonance frequencies, i.e. ω_1, ω_2 and ω_3, can be regarded as the isolated qubits ($\omega_1 < \omega_2 < \omega_3$).

In this experiment, the difference frequencies of the three spins are $\omega_3 - \omega_2 = 471.8Hz$ and $\omega_3 - \omega_1 = 700.3Hz$. The corresponding J coupling constant between the two qubits are $J_{13} = 10.9Hz$, $J_{23} = 17.6Hz$, and $J_{12} = 0.65Hz$. The transmitter frequency of the transition-pulse is set at the $\omega_3/2\pi + J_{13}/2 + J_{13}/2$. The power and the duration of the transition-pulse are adjusted to optimize the frequency selectivity. The real part of the Fourier transform of the resulting signal gives a NMR spectrum of the target qubit-3 containing four peaks at frequencies $\omega_3/2\pi \pm J_{13}/2 \pm J_{13}/2$. these four peaks (from left to right) correspond to the two control qubits in the following states $|00\rangle$, $|01\rangle$, $|10\rangle$ and $|11\rangle$. It is clear that our implementation of the quantum $\Lambda_2\,(not)$ gate leaves the computer in a final result as expected: if any one of the two control qubits is in ground state , the target qubit-3 does not change its value after the action of the gate; and if the two control qubits are both in states $|1\rangle$, the target qubit-3 flips its state $|0\rangle \leftrightarrow |1\rangle$.

B. four-qubit $\Lambda_3\,(not)$ gate

The experimental realization of a modified four-qubit quantum $\Lambda_3\,(not)$ gate is shown in Figure 2. The physical system that we used to implement the $\Lambda_3\,(not)$ gate is the liquid-state proton and carbon NMR of $^{13}C_3$ labeled alanine $NH_3^+ - C^\alpha H\left(C^\beta H_3\right) - C^i OOH$, with decoupling of the methyl protons, alanine exhibits a weakly coupled four-spin system. The C^α was chosen as target qubit (labeled qubit-4) because of its well-resolved couplings with the other carbons $\left(J_{C^\alpha C^i} = 53.92Hz,\ J_{C^\alpha C^\beta} = 34.42Hz\right)$ and with the adjacent proton ($J_{C^\alpha H} = 144.89Hz$), which were chosen as the three control spins (labeled qubit-1, qubit-2 and qubit-3). The selective π pulse were applied on the connected single-quantum transition $|1_1 1_2 1_3 0_4\rangle \leftrightarrow |1_1 1_2 1_3 1_4\rangle$ of the target qubit-4, it is noted that all eight lines arise from the target qubit-4, and the intensity of each line is proportional to the total population difference between the corresponding states. It is clear that our implementation of the quantum logic gate leaves the computer in a final result as expected,

All NMR experiments are performed at room temperature and pressure on Bruker Avance $DMX500$ spectrometer ($11.7T$) with a $5mm$ probe in Laboratory of Structure Biology, University of Science and Technology of China. We can see the small but significant distortions in the final spectra arising from the difficulty of implementing perfect selective pulses and inhomogeneity of the magnetic field. The experimental task is to shape the radio-frequency pulse envelope so as to achieve sufficient selectivity in the frequency domain that there is negligible perturbation of the next nearest neighbor of the spin multiplet.

IV. CONCLUSION

In conclusion, utilizing the proper resonant pulse of the well-defined frequency and length, we have demonstrated the reversible and universal three and four-qubit quantum $\Lambda_n\,(not)$ logic gates in two-level nuclear spin systems at room temperature.

One of the key challenges in quantum computation is to try to increase the size of the system used. The transition pulse that used in this paper can apply to all qubits of a quantum register in one operation. the operational time of the pulse is not influenced by the number of the qubits, therefore, it is experimentally convenient when we extend this procedure to more qubits. This advantage may be helpful to answer the question given a unitary transformation corresponding to a quantum computational task, what is the shortest sequence of pulses and evolutions which generates it? Further, in quantum information processing, this single-pulse quantum operation is useful not only in an ensemble of nuclear spins system but also in many other physical systems such as quantum dots, trapped ions *etc.*

Although the multi-qubit gates implementation with transition pulse is much simpler, it must be noted that it takes more time than the multi-qubit gates implementation with spin-selective pulse. For the experimenters, a reasonable choice should be to combine the utility of various NMR methods.

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Figure Captions:

Fig. 1, The Λ_3 (*not*) quantum gate implemented on the four-spin system of alanine. (*a*) experimental NMR spectra of the target qubit of the input thermal equilibrium state. (*b*) experimental NMR spectra of the target qubit of the output state of the Λ_3 (*not*) gate.

Fig. 2, The Λ_3 (*not*) quantum gate implemented on the four-spin system of alanine. (*a*) experimental NMR spectra of the target qubit of the input thermal equilibrium state. (*b*) experimental NMR spectra of the target qubit of the output state of the Λ_3 (*not*) gate.
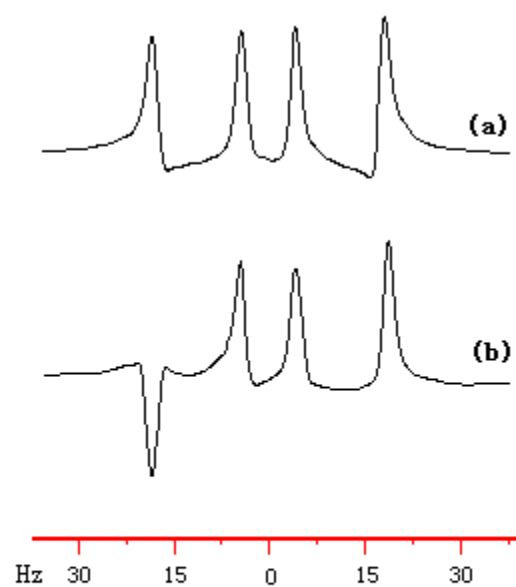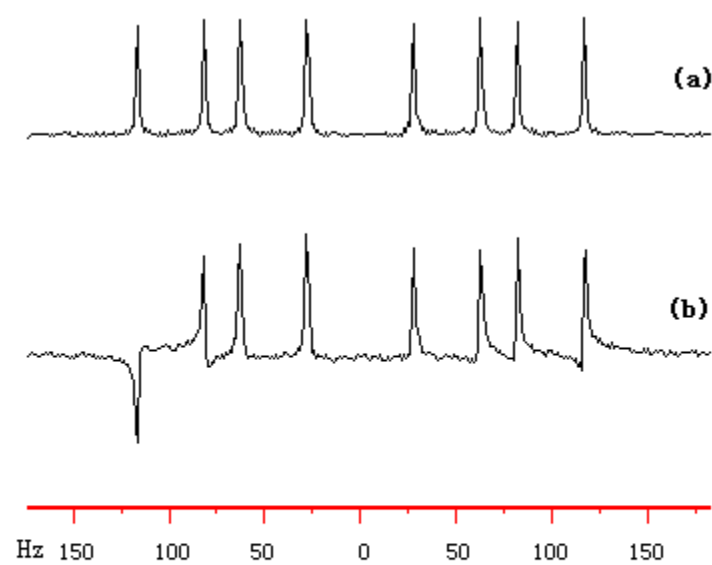
Hz 30   15   0   15   30

Fig. 1, Du

(a)

(b)

Hz 150   100   50   0   50   100   150

Fig. 2